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TRANSURANIUM PROCESSING PLANT SEMIANNUAL REPORT OF PRODUCTION, STATUS, AND PLANS FOR PERIOD ENDING JUNE 30, 1968

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SUMMARY

This report introduces a series of progress reports that will be issued semiannually to inform the heavy-element community of the status and the future production plans of the Transuranium Element Production Program at ORNL. The objective of these reports is to provide information that will enable users of the products to obtain the maximum service from the production facilities at ORNL. Production plans and schedules are definitely established only for the short term; long-range plans can be (and are) markedly influenced by feedback from researchers. This first report summarizes the project to date (covering a period of about two years); subsequent reports will summarize the events of the preceding six months.

The Transuranium Processing Plant (TRU) and the High Flux Isotope Reactor (HFIR) have been built at ORNL to produce gram quantities of many of the transuranium elements, and milligram quantities of some of the transcalifornium isotopes, for use in research work by laboratories throughout the country.

The total amounts of transuranium elements processed during the first two-year period are (approximately): 125 g of ^{242}Pu, 170 g of ^{243}Am, 300 g of ^{244}Cm, 1 mg of ^{249}Bk, 7 mg of ^{252}Cf, and 40 μg of ^{253}Es. Approximately 91 g of ^{242}Pu, 207 g of ^{243}Am, 239 g of ^{244}Cm, 25 μg of ^{249}Bk, and 1.7 mg of ^{252}Cf are still in various stages of purification in TRU. We have made about 74 shipments, which have totaled: 0.8 mg of ^{244}Pu, 37 g of ^{243}Am, 79 g of ^{244}Cm, 36 μg of ^{248}Cm, 411 μg of ^{249}Bk, 72 μg of ^{249}Cf (isotopically pure), 1 mg of ^{252}Cf, 1 μg of ^{253}Es (isotopically pure), and 16 μg of ^{253}Es containing some ^{254}Es and ^{255}Es.

We anticipate the production of about 5.5 mg of ^{249}Bk, 50 mg of ^{252}Cf, 260 μg of ^{253}Es, and 10^9 atoms of ^{257}Fm during the next eighteen months. A total of about 1 g of californium is expected to be produced by about 1973.

The processes and equipment that are used in TRU are being changed constantly as our knowledge increases. The current sequence of process steps is presented and the status of each step is summarized. The steps are as follows: (1) a feed solution is prepared by dissolving irradiated targets; (2) the plutonium is recovered using a batch solvent extraction process (Pubex); (3) the transplutonium elements are decontaminated from

fission products by countercurrent solvent extraction (Tramex); (4) the americium and curium are separated from the transcurium elements by ion exchange; (5) the transcurium elements are separated from each other by chromatographic elution with α-hydroxyisobutyrate from a high-pressure ion exchange column; (6) the plutonium, americium, and curium are made into oxide microspheres, using a sol-gel technique; and (7) the microspheres are fabricated into HFIR targets, by remote means, for reirradiation to produce more transcurium elements.

Sixteen TRU target rods developed longitudinal cracks in the cladding after irradiation in the HFIR for 125 to 175 days at a power level of 100 Mw. They were part of a group of 17 targets that had been irradiated for about one year in a Savannah River Plant reactor. A mathematical model, formulated to guide the development of an improved target design, indicates that a smaller loading of plutonium per target and/or pressing the pellets to lower densities will result in longer target lifetimes. Test targets are now being irradiated to confirm this.

The services available at TRU have been used to prepare special materials that are not normally produced in main-line efforts. Three special californium targets were fabricated and irradiated to produce einsteinium and to refine some of the calculated values of cross sections in the chain of isotopes above californium. Aluminum tensile specimens were irradiated for use in studying the target failure problem. About 350 μg of ^{252}Cf was loaded into a small aluminum capsule for use as a source in fast-neutron activation studies. Uranium oxide ($^{236}U_3O_8$) was fabricated into pellets and irradiated in the HFIR hydraulic rabbit facility for the production of ^{237}U. The rabbits were subsequently processed by personnel at the Los Alamos Scientific Laboratory, and the uranium was used in an underground nuclear event for the measurement of the fission cross section of ^{237}U. Rabbits containing ^{253}Es were fabricated and were irradiated in the HFIR to produce 39.3-hr ^{254m}Es for studies at the Argonne National Laboratory.

The values that we use for transuranium-element decay data and for cross-section data in planning irradiation-processing cycles, calculating production forecasts, and assaying products are tabulated in the Appendix.

1. INTRODUCTION

This report introduces a series of progress reports that will be issued semiannually to inform the heavy-element community of the status of the Transuranium Element Production Program at ORNL and of future production plans. The objective of these reports is to provide information that will enable users of the products to obtain the maximum service from the production facilities at ORNL. Production plans and schedules are definitely established only for the short term; long-range plans can be (and are) markedly influenced by feedback from researchers. In this first report we will summarize the project to date (covering a period of about two years); in subsequent reports we will be concerned only with the events of the preceding six months. Transuranium Quarterly Progress Reports[1] were published from 1962 through 1965 to record the development of separation processes for the transuranium elements, the development of process equipment, the development of methods for fabricating HFIR targets, the progress in the construction of the Transuranium Processing Plant, the results of corrosion studies, and the data accumulated from analytical chemistry research and development. Descriptions of the Transuranium Processing Plant and proposed process flowsheets and operating techniques are included in the Safety Analysis.[2]

The Transuranium Processing Plant (TRU) and the High Flux Isotope Reactor (HFIR) have been built at ORNL to produce gram quantities of many of the transuranium elements, and milligram quantities of some of the transcalifornium isotopes, for use in research work by laboratories throughout the country. This part of the USAEC Heavy Element Production Program began with the long-term irradiation of 10-kg batches of ^{239}Pu in the Savannah River Plant (SRP) reactors. About 1 kg of ^{242}Pu and about 300 g each of ^{243}Am and ^{244}Cm were produced. The ^{242}Pu was recovered and decontaminated at SRP, and most of the americium and curium was stored at ORNL for further processing. A portion of the ^{242}Pu was fabricated into target rods that are now being irradiated in the HFIR. To accelerate the production rate of transuranium elements while TRU and HFIR were being built, about 525 g of the ^{242}Pu was irradiated during a

special High Flux Demonstration Run[3] in one of the SRP production reactors. The 525 g of ^{242}Pu was fabricated into three forms: (1) eight slugs of usual geometry for the SRP reactor; (2) six prototype HFIR targets, which contained 10 g of $^{242}PuO_2$ each; and (3) 18 actual HFIR target rods, which also contained 10 g of $^{242}PuO_2$ each.

The first TRU processing campaign, which began in August 1966 and was completed in November 1966, was concerned with the prototype HFIR targets.[4] The slugs that had been irradiated in the SRP reactor[5] were processed next. The actual HFIR targets, which had been irradiated at the Savannah River plant for about one year, began to fail after about 125 days of irradiation in the HFIR. When all except one had failed, the entire group was processed in TRU for the recovery of the heavy elements. About one-half of the americium-curium-bearing raffinate from the SRP plutonium recovery processing has now been recovered and purified in TRU.

This report summarizes TRU operations to date and specifies the quantities of materials that have been produced, processed, and shipped. We have presented proposed processing schedules and anticipated yields of various products, along with a discussion of the mathematical models that were used for making the calculations. The status of the development of each proposed processing step (such as Tramex processing or target fabrication) is briefly discussed, and major problem areas are enumerated. Special processing, fabrication, and irradiation programs are described. The Appendix contains a tabulation of the values of nuclear parameters which were used as input data for the calculations of production rates for transuranium elements, and a tabulation of the parameters which were used to calculate the specific activities of the isotopes that are of interest to TRU.

2. PROCESSING SUMMARY AND PRODUCTION ESTIMATES

The isotopic concentrations of the various transuranium elements are not constant, but are functions of irradiation histories and decay times. We use the major isotope in a mixture of isotopes to determine

material balances. Thus we trace curium by the isotope ^{244}Cm and we trace californium by the isotope ^{252}Cf. Except in special instances, ^{242}Pu, ^{243}Am, ^{249}Bk, and ^{253}Es are the isotopes used for tracing these elements. Throughout this report section we are discussing mixtures of isotopes when we do not stipulate "isotopically pure."

2.1 Processing Summary

The following total amounts (approximate) of transuranium elements were processed during the first two-year period of TRU operations through June 30, 1968: 125 g of ^{242}Pu, 170 g of ^{243}Am, 300 g of ^{244}Cm, 1 mg of ^{249}Bk, 7 mg of ^{252}Cf, and 40 μg of ^{253}Es. The amounts processed in each of the major campaigns are given in Table 2.1. The amounts of short-lived nuclides were corrected to the date of each dissolution, and were summed for all dissolutions without further adjustment to a common time basis.

Seventy-four shipments have been made from TRU (see Table 2.2). These shipments included:

(1) 0.8 mg of ^{244}Pu that had been isotopically enriched in calutrons and then irradiated in the HFIR prior to shipment;

(2) 36.9 g of ^{243}Am and 78.7 g of ^{244}Cm that had been separated and purified (we do not usually separate americium and curium);

(3) 36 μg of ^{248}Cm that had been "milked" from californium;

(4) 411 μg of ^{249}Bk;

(5) 72.4 μg of ^{249}Cf (isotopically pure) that had been "milked" from ^{249}Bk;

(6) 1.1 mg of ^{252}Cf in a mixture of californium isotopes (75-80% ^{252}Cf);

(7) 15.5 μg of ^{253}Es containing some ^{254}Es and ^{255}Es;

(8) 1.1 μg of ^{253}Es (isotopically pure) that had been "milked" from californium.

As of June 30, 1968, our inventory of transuranium materials was 91 g of ^{242}Pu, 207 g of ^{243}Am, 239 g of ^{244}Cm, 25 μg of ^{249}Bk, and 1.7 mg of ^{252}Cf. Typical isotopic concentrations of the curium and californium are given in Table 2.3.

Table 2.1. Amounts of Materials Processed During Major Campaigns in the Transuranium Processing Plant Prior to July 1,1968

Listed values are measurements or estimates of feed solutions

Campaign No.	Completion Date	Campaign Description	^{242}Pu (g)	^{243}Am[a] (g)	^{244}Cm[a] (g)	^{249}Bk (μg)	^{252}Cf (mg)	^{253}Es (μg)
1	July 1966	Tracer-level tests	b	b	b	b	b	b
2	November 1966	Four prototype HFIR targets	10.5	4.5	12.4	30[c]	0.1[d]	b
3	April 1967	Six SRP reactor slugs	49.5	19.3	76.5	440[c]	2.3	14.2
4	May 1967	Five ruptured HFIR targets	12.0	5.7	22.1	170[c]	1.0	3.4
5	July 1967	Miscellaneous targets[e]	26.5[d]	7.7[d]	17.0	b	0.1[d]	b
6	August 1967	Twelve ruptured HFIR targets	26.3	15.2	46.4	475[c]	4.0	23.3
7	Not Completed[f]	Am-Cm solution from SRP	b	118.7	131.5	b	b	b
8	May 1968	Rework material from other campaigns	b	(14)	(31)	(80)	(0.7)	(b)
		Total (excluding rework)	124.8	171.1	305.9	1115	7.4	40.9

[a]Americium and curium are not usually separated from each other. About 80 g of ^{244}Cm and 40 g of ^{243}Am were separated for special programs.

[b]Amounts were known to be trivial.

[c]Analytical method for berkelium in feed solutions was not available. Listed values were estimated from measurements of ^{248}Cm.

[d]Estimated from product measurements.

[e]Six targets were dissolved in laboratory-size equipment to obtain more-precise data.

[f]Continuing at present.

Table 2.2. Distribution of Heavy Elements from the Transuranium Processing Plant Through June, 1968

Major Nuclide	Date	Shipped To Individual	Shipped To Site
Plutonium-244 (irradiated), mg			
0.5	3-13-68	J. Halperin	ORNL
0.3	3-13-68	H. Diamond	ANL
0.8			
Americium-243, g			
13.5	4-11-67	R. A. Penneman	LASL
0.2	5-10-67	O. L. Keller	ORNL-TRL
0.1	6-09-67	Isotopes Sales	ORNL
1.0	9-08-67	O. L. Keller	ORNL-TRL
19.6	4-01-68	R. A. Penneman	LASL
0.5	4-05-68	R. M. Latimer	LRL-B
2.0	4-05-68	R. W. Hoff	LRL-L
36.9			
Curium-244, g			
2.1	3-23-67	R. W. Hoff	LRL-L
10.0	4-04-67	W. D. Box	ORNL
3.3	9-06-67	R. W. Hoff	LRL-L
38.1	2-21-68	T. A. Butler	ORNL
1.7	3-21-68	W. D. Box	ORNL
0.9	3-28-68	J. L. Burnett	ORNL-TRL
9.4	4-12-68	R. A. Penneman	LASL
2.1	4-12-68	M. S. Moore	Idaho Nuclear Co.
2.1	5-02-68	R. O. Budd	PNL
6.6 (low Am)	6-10-68	B. Rushe	SRL
2.4	6-10-68	R. D. Kelsch	SRL
78.7			
Curium-248, μg			
10	1-22-68	R. Folger	SRL
13	5-28-68	C. Bemis	ORNL-TRL
13	6-17-68	C. Bemis	ORNL-TRL
36			

Table 2.2 (continued)

Major Nuclide	Date	Shipped To Individual	Shipped To Site
Berkelium-249, μg			
70	10-11-67	R. D. Baybarz	ORNL
70	10-11-67	A. Ghiorso	LRL-B
70	10-11-67	R. W. Hoff	LRL-L
70	10-11-67	R. A. Penneman	LASL
70	10-11-67	P. R. Fields	ANL
4	11-14-67	J. Halperin	ORNL
1	5-16-68	B. Weaver	ORNL
8	5-20-68	P. Fields	ANL
8	5-20-68	R. W. Hoff	LRL-L
8	5-20-68	T. C. Parsons	LRL-B
8	5-20-68	M. L. Hyder	SRL
16	6-14-68	R. Baybarz	ORNL
8	6-14-68	R. Baybarz	ORNL
411			
Californium-249 (isotopically pure), μg			
55	9-28-67	A. Ghiorso	LRL-B
7.4	1-16-68	J. Halperin	ORNL
10	5-16-68	A. Ghiorso	LRL-B
72.4			
Californium-252, μg			
50	1-26-67	P. R. Fields	ANL
50	1-26-67	A. R. Boulogne	SRL
3	6-01-67	O. L. Keller	ORNL-TRL
10	8-07-67	O. L. Keller	ORNL-TRL
100	9-08-67	A. R. Boulogne	SRL
10	9-15-67	Isotopes Sales	ORNL
10	11-14-67	J. Halperin	ORNL
200	12-08-67	C. H. Youngquist	ANL
100	12-15-67	A. R. Boulogne	SRL
10	1-16-68	Isotopes Sales	ORNL
25	1-22-68	A. R. Boulogne	SRL
350	2-12-68	Enzo Ricci	ORNL
10	4-01-68	Isotopes Sales	ORNL
100	4-12-68	R. A. Penneman	LASL

Table 2.2 (continued)

Major Nuclide	Date	Shipped To Individual	Shipped To Site
Californium-252 (continued), μg			
50	4-12-68	M. S. Moore	Idaho Nuclear Co.
1	4-23-68	C. Bemis	ORNL-TRL
1	4-30-68	E. K. Hulet	LRL-L
11	5-08-68	C. Bemis	ORNL-TRL
1	5-15-68	R. L. Folger	SRL
50	6-10-68	A. R. Boulogne	SRL
1142			
Einsteinium-253 (isotopically pure), μg			
1.0	5-13-68	H. Diamond	ANL
0.1	6-14-68	H. Diamond	ANL
1.1			
Einsteinium-253, μg			
1	5-19-67	E. K. Hulet	LRL-L
2	6-09-67	B. B. Cunningham	LRL-B
0.1	7-01-67	R. A. Penneman	LASL
0.7	7-01-67	P. R. Fields	ANL
3.2	9-15-67	P. R. Fields	ANL
2.8	9-22-67	P. R. Fields	ANL
0.6	3-18-68	T. C. Parsons	LRL-B
0.8	4-05-68	T. C. Parsons	LRL-B
1.4	4-05-68	R. W. Hoff	LRL-L
1.4	4-05-68	P. R. Fields	ANL
0.5	4-12-68	T. C. Parsons	LRL-B
0.5	4-12-68	R. W. Hoff	LRL-L
0.5	4-12-68	P. R. Fields	ANL
15.5			

Table 2.3. Isotopic Concentrations of Some of the Curium and Californium in TRU on July 1, 1968

Californium

Isotope	Atom %
^{249}Cf	4
^{250}Cf	16
^{251}Cf	5
^{252}Cf	75

Curium

	Atom %	
Isotope	From Irradiated Targets	From SRP Solution
^{242}Cm		0.002
^{243}Cm		0.037
^{244}Cm	90.89	94.66
^{245}Cm	0.769	1.770
^{246}Cm	7.87	3.449
^{247}Cm	0.200	0.063
^{248}Cm	0.266	0.019

2.2 Irradiation and Processing Proposals

The estimated future production rates of transcurium elements are given in Table 2.4. Because of its relatively long half-life, ^{252}Cf is used to indicate long-term predictions; ^{249}Bk and ^{253}Es are listed for only the next 18 months. The estimate of californium production is based on the assumption that 80% of all of the curium isotopes that are recovered during each processing campaign are recycled to the HFIR for transcurium element production. The other 20% has been assumed to constitute product shipments (probably after mass separation in the calutrons to concentrate heavy isotopes) and processing losses.

In July 1968 we plan to process 13 plutonium targets that have been irradiated in the HFIR since it first went to power in September 1966. Each target originally contained 8 g of plutonium (95.7% ^{242}Pu) as PuO_2. About 1 mg of ^{249}Bk, 6 mg of ^{252}Cf, and 30 μg of ^{253}Es will be purified as products, and 15 g of ^{242}Pu, 5 g of ^{243}Am, and 40 g of ^{244}Cm will be recovered for use in making recycle targets. Then we plan to recover about 120 g each of ^{244}Cm and ^{243}Am, from the remaining SRP raffinate solution plus some rework material, to provide feed material for future irradiation. We expect to isolate an interesting californium product (300 μg) from the SRP raffinate solution. Because it will contain only about 20% ^{252}Cf it will emit much less hazardous radiation per unit mass than our usual californium product. Therefore, radiochemists can experiment with four to five times as much of this californium as ^{252}Cf. Since it contains about 25% ^{250}Cf, this californium will also be of interest as a target material for heavy ion bombardment.

Six americium-curium targets will be processed in January 1969 after having been irradiated for seven months. This will be the first group of americium-curium targets (see Sect. 3.8). Each target originally contained 4.4 g of ^{244}Cm, 0.3 g of heavier curium isotopes, and 0.8 g of ^{243}Am. We expect to recover 1.2 mg of ^{249}Bk, 10 mg of ^{252}Cf, and 47 μg of ^{253}Es as purified products.

Also, in the early part of 1969, we expect to obtain about 20 μg of einsteinium from the irradiation of 4 mg of ^{252}Cf in the HFIR hydraulic

Table 2.4. Estimated Future Production of Transcurium Elements

Period	Processing Campaign	Products of Campaigns			^{252}Cf Production		Date Products Available
		^{249}Bk (mg)	^{252}Cf (mg)	^{253}Es (μg)	Annual (mg)	Cumulative (mg)	
Through June 1968						5	
July - December 1968	13 Pu targets[a]	1	6	30		11	August 1968
1969	6 Cm targets (second cycle)	1.2	10	47			March 1969
	Hydraulic rabbit (Cf irradiation)			20			March 1969
	7 Pu targets[a]	1.3	7	35			August 1969
	3 Cm targets (third cycle)[b]	2	28	132	45	56	December 1969
1970					145	201	
1971					300	501	
1972					440	941	
1973					570	1511	

[a]Each of the targets originally contained 8 g of plutonium (95.7% ^{242}Pu) as PuO_2.

[b]These targets will contain unburned curium from the 6 Cm targets listed above.

rabbit facility. We produced some einsteinium in March 1968 by irradiating approximately 3 mg of ^{252}Cf in targets in the HFIR flux trap (Sect. 4.1); however, the yield was disappointingly low (only 6 μg as compared with the 18 μg expected) because neutron cross-sections for the reactions involved had been incorrectly estimated. In the rabbit facility we will irradiate the ^{252}Cf for a short time to produce ^{253}Cf and then withdraw it to allow the ^{253}Cf to decay to ^{253}Es. We plan 3 cycles of 3 days of irradiation and 15 days of decay. Based on our estimates of the parameters for the competing reactions, this is the optimum production schedule.

In July 1969, seven plutonium targets (8 g of Pu - 95.7% ^{242}Pu) will be processed to recover about 1.3 mg of ^{249}Bk, 7 mg of ^{252}Cf, and 35 μg of ^{253}Es as products, and 6 g of ^{242}Pu, 2.5 g of ^{243}Am, and 20 g of ^{244}Cm for recycle to the HFIR.

About 13 g of ^{244}Cm and 2.6 g of heavier curium isotopes (80% of the curium recovered in January from the six americium-curium targets) will be fabricated into three targets which will be irradiated for six months and will then be processed in the latter part of 1969. About 2 mg of ^{249}Bk, 28 mg of ^{252}Cf, and 132 μg of ^{253}Es will be recovered as purified products; the residual curium will be recycled to the HFIR for additional irradiation.

2.3 Estimates of the Availability of Transuranium Elements

Although plutonium, americium, and curium are usually considered to be intermediate feed materials rather than products, we have purified and distributed isotopes of these elements (primarily ^{242}Pu, ^{243}Am, and ^{244}Cm). We also expect to provide small quantities of the heavy curium isotopes, ^{246}Cm, ^{247}Cm, and ^{248}Cm, for experimental work. However, these isotopes are very fertile feed materials for the production of transcurium elements, and the production rate of transcurium elements will be drastically reduced if a large fraction of the available heavy curium isotopes is removed from the production line.

2.3.1 Berkelium

About 5.5 mg of ^{249}Bk will be available during the next 18 months: 1 mg in August 1968, 1.2 mg in March 1969, 1.3 mg in August 1969, and 2 mg in December 1969. It would be possible to increase the rate of production of berkelium if irradiation-processing schedules were optimized for the production of berkelium instead of californium. However, this would considerably decrease the availability of californium.

2.3.2 Californium

We expect to produce 6 mg of mixed californium isotopes (about 75-80% ^{252}Cf) in August 1968 and 45 mg in 1969. In the last half of 1968, about 300 μg of californium will be recovered from the SRP raffinate solution. This product will contain about 45% ^{249}Cf, 25% ^{250}Cf, 10% ^{251}Cf, and 20% ^{252}Cf.

Production of isotopically pure ^{249}Cf by "milking" ^{249}Bk is not anticipated in the next 18 months because we expect to ship all of the berkelium as soon as it becomes available.

2.3.3 Einsteinium

We will recover 30 μg of ^{253}Es (about 0.3% ^{254}Es and 0.06% ^{255}Es) in August 1968. In March 1969 we expect to recover 47 μg from targets and 20 μg from the irradiation of 4 mg of californium in the HFIR hydraulic rabbit facility. About 35 μg will be separated in about August 1969, and 132 μg is anticipated from three curium targets (third-cycle) that will be processed about December 1969.

After the mixture of einsteinium isotopes has been separated from it, the purified californium will be stored to allow ^{253}Es to be formed from the decay of ^{253}Cf, and the second-growth ^{253}Es will be recovered. About 17 μg of isotopically pure ^{253}Es will be "milked" from the californium that will be isolated in March 1969, and 9 μg will be milked from the californium that will be isolated in August 1969.

2.3.4 Fermium

Approximately 1.5 x 10^7 atoms of ^{257}Fm per milligram of ^{252}Cf will be present in each batch of californium that will be recovered. Thus, about 9 x 10^7 atoms will be available in August 1968, 2 x 10^8 atoms in March 1969, 1 x 10^8 atoms in August 1969, and 4 x 10^8 atoms in December 1969.

2.4 Computational Methods and Nuclear Cross Sections

Two computer programs are used in predicting the compositions of targets that have been irradiated in the HFIR. The first program, an extension of earlier programs based on the CRUNCH[6] code, is used to calculate production rates of the various isotopes during neutron irradiation. It has two distinguishing features: it includes explicit contributions from epithermal neutron reactions, and it makes an approximate calculation of resonance self-shielding effects, which cause the effective neutron cross sections for particular nuclides to vary during irradiation as the concentrations of these nuclides change.

Two ^{242}Pu targets, which were irradiated at the SRP to about 45% burnup and were subsequently analyzed for heavy-element content, served as the first subjects for this code. Numerous calculations were made using the known irradiation histories of these targets and various assumed values for thermal-neutron cross sections. The results of these calculations were then analyzed, using the second computer program, which interpolates and weighs the results by the least-squares method, to obtain the set of cross sections that most nearly results in the composition observed at the end of irradiation. This second program is capable of simultaneously considering the data for ten different targets.

In the analysis to date, resonance absorption has been based on literature values of infinite-dilution resonance integrals (1280 and 1500 barns for ^{242}Pu and ^{243}Am, respectively). For the purpose of estimating resonance self-shielding, a mathematical model that is appropriate to a single narrow resonance was assumed. The values for thermal

cross sections (2200 m/sec) resulting from this analysis agree with the lower end of the range of values reported in the literature (see Table 2.5).

The results of the analysis of the SRP irradiations were used to predict the HFIR effective cross sections of ^{242}Pu and ^{243}Am, the first two members of the transuranium-element production chain (Table 2.6). Reactor effective cross sections vary during irradiation as target compositions change; this analysis was based on the target compositions at the end of one reactor-core lifetime in the HFIR.

HFIR target D-30, which had been irradiated for one reactor-core cycle at 90 Mw, was dissolved, and the resulting solution was analyzed for transuranium elements. The results were then subjected to numerical analysis. Estimates of HFIR effective cross sections for ^{242}Pu and ^{243}Am are within 10% of the values previously calculated. This agreement is encouraging, indicating that there are no gross deficiencies in the mathematical model that is used to allow for resonance self-shielding for these two isotopes.

Predictions of the ^{242}Pu, ^{243}Am, and ^{244}Cm contents of the first batch of 17 targets irradiated both at the SRP and in the HFIR were within 10% of the measured values. The predicted content of ^{252}Cf was too high by a factor of 1.8.

As larger quantities of the transuranium elements are produced, these two programs will be used repeatedly to reevaluate cross-section data and to make better predictions of the availability of various isotopes.

3. PROCESSES AND EQUIPMENT

The processes and equipment that are used in TRU are constantly being modified as our knowledge increases. These modifications range from trivial changes (i.e., changing the concentration of a process stream) to significant ones (i.e., using a completely new process to perform one of the processing steps).

Table 2.5. Equivalent 2200-m/sec Cross Sections[a]

	^{242}Pu	^{243}Am
1. SRP slug	19.12	83.56
2. HFIR prototype	18.04	63.93
3. Combined analysis of 1 and 2	18.57	74.85
4. HFIR-irradiated target D-30	16.39	73.66
5. Combined analysis of 1, 2, and 3	18.00	73.74
Literature values[b]	17.5-30	73.6-183

[a]In barns.
[b]Cross Sections for Transuranium Element Production, BNL-982 (T-415), Neutron Cross Section Evaluation Group, Brookhaven National Laboratory (May 1966).

Table 2.6. HFIR Effective Cross Sections[a]

	Cross Section (barns)		
	Predicted (from SRP Data)	Observed (D-30)	Percentage Difference
^{242}Pu	22.1	20.3	-8.3
^{243}Am	99.6	98.6	-1.0

[a]Reactor effective cross sections vary during irradiation as target compositions change. These values are for the end of one irradiation cycle.

Because this is the first report in a series, we will summarize the status of process development for each process step and will discuss the most important changes that have been made since TRU became operational. Also, we will discuss some of the major problems that we have encountered.

Fig. 3.1 is a block diagram showing the processing steps that are required for transuranium element production. These steps are: (1) the preparation of a feed solution by dissolving irradiated targets; (2) the recovery of plutonium; (3) the decontamination of the transplutonium elements; (4) the separation of americium and curium from the transcurium elements; (5) the separation and purification of berkelium, californium, einsteinium, and fermium; (6) the preparation of americium-curium oxide microspheres; and (7) the fabrication of targets to be irradiated in the HFIR.

The first four processing steps, plus target fabrication, are being performed routinely in the main TRU operating facilities. Plant-scale equipment for making americium-curium oxide microspheres is being installed. Plant-scale processes (and equipment) for separating and purifying the transcurium elements are still in the development stage; in the interim, the separation and purification are being carried out in equipment that is installed in cell 5 in TRU. When a process becomes sufficiently developed, equipment for it will be installed in the main-line processing area; the temporary equipment will then be removed from cell 5.

Product finishing operations and special separations will continue to be performed in cell 5 and in supporting shielded cave facilities. As products reach certain levels of purity, they must be moved to progressively cleaner facilities and finally to shielded caves or to glove boxes that are used only for handling products.

3.1 Description of a Target

A HFIR target rod (see Fig. 3.2) consists of actinide-bearing compacts (pellets) with inner containment jackets, an outer cladding tube

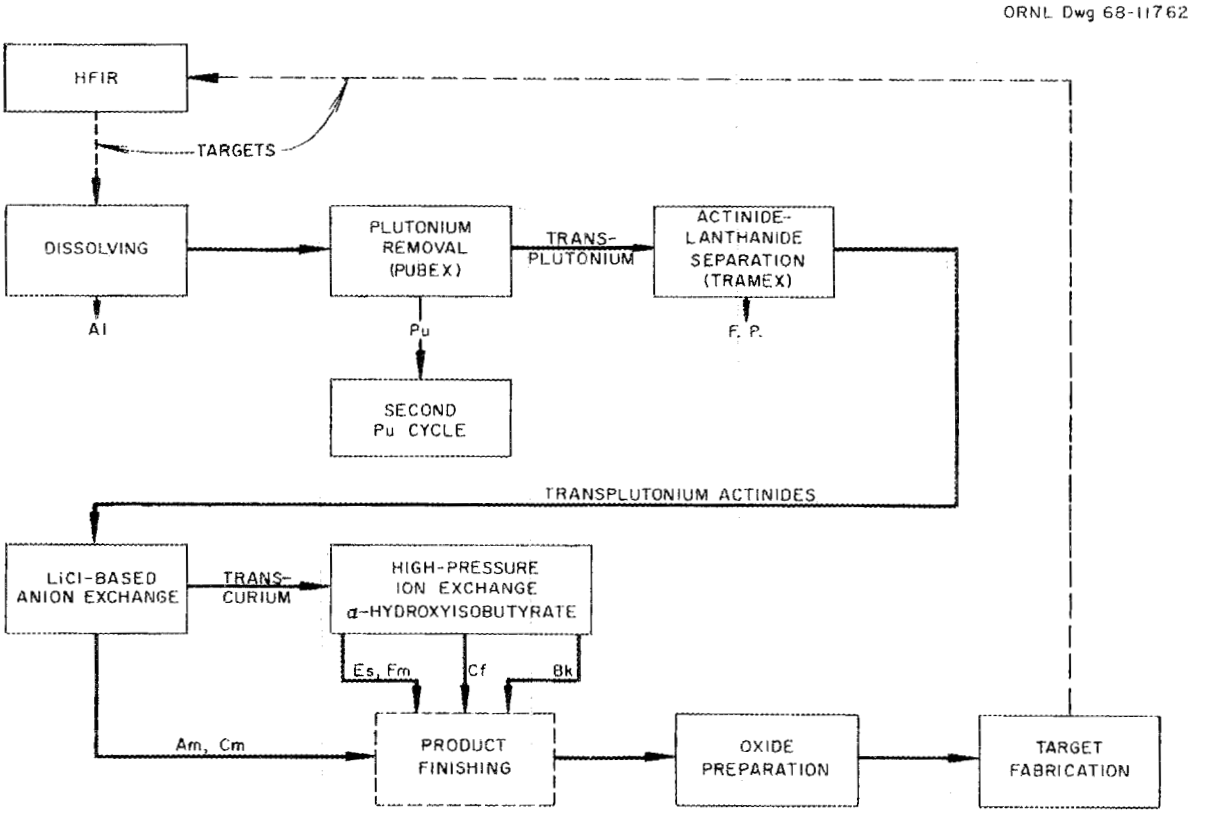

Fig. 3.1. Processing Steps Used for Transuranium Element Production.

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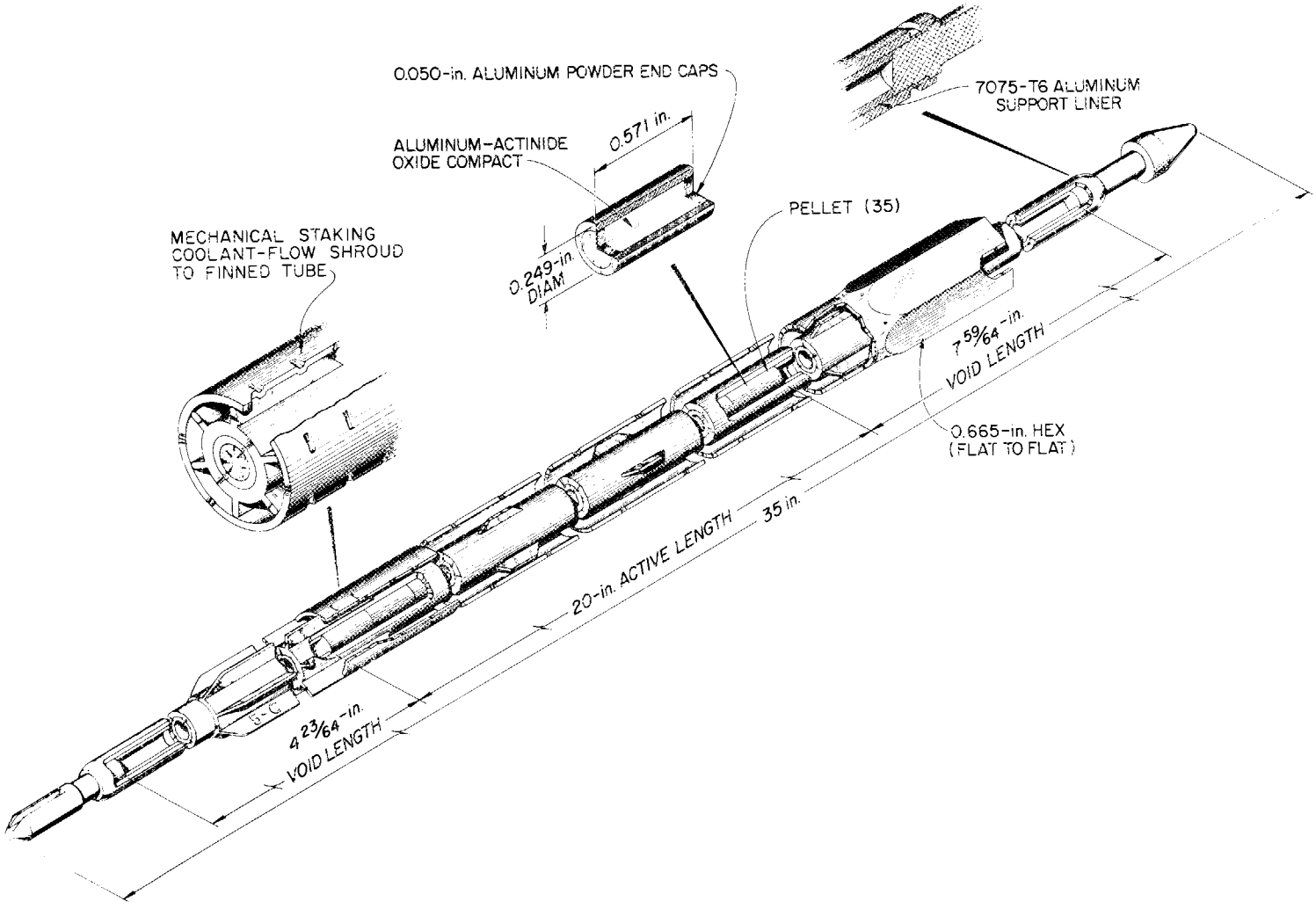

Fig. 3.2. HFIR Target Rod.

with intermittent fins, support liners and plugs, and an outer spacing sheath (coolant-flow shroud tube). Usually, 35 aluminum-clad pellets containing actinide oxides dispersed in aluminum powder are included in the 20-in. active length. These pellets are nominally 0.249 in. in diameter and 0.571 in. long. The pellets and the support liners are sealed in the target tube by welded end plugs. The hex sheath for coolant flow is mechanically staked to intermittent fins on the target tube.

3.2 Target Dissolution

The main-line process for dissolving targets is routine in TRU. The aluminum coolant-flow shroud tube is removed mechanically, and the remaining aluminum (target rod and matrix) is dissolved in a Zircaloy-2 dissolver using 6 M NaOH--3 M $NaNO_3$. The aluminum-bearing solution is decanted through a filter, and the residue of actinide oxides is dissolved using 6 M HCl. Twenty targets have been dissolved in this manner. No trouble has been experienced in retaining the actinide residue while removing the caustic solution; less than 0.02% of the curium has been lost to the dejacketing waste.

There are two advantages of caustic dejacketing: (1) Removing the aluminum prior to Tramex increases the throughput rate of actinides about tenfold (because of the low solubility of aluminum in Tramex feed solution). (2) Much of the radioiodine is retained by the caustic solution, thereby allowing the processing of short-cooled targets.

Some plutonium oxide targets in which the burnup of plutonium is less than 70 to 75% require fluoride-catalyzed HNO_3 for complete dissolution. The prototype HFIR targets and some of the SRP slugs were dissolved with this reagent in a temporary stainless steel dissolver system, which has been removed from the cells. Then, a fluoride removal treatment was required because the TRU equipment, which is fabricated from Zircaloy-2 or tantalum, is not compatible with fluoride. We believe that very little low-burnup material will have to be processed in the future. Thus it can be dissolved and treated for fluoride removal in small, glass equipment prior to being introduced into main-line equipment.

3.3 Recovery of Plutonium

When TRU first became operational, we expected to use an anion exchange method (the Plurix process) to recover plutonium. The nitrate-based version of the Plurix process, which has been in use for many years, proved to be satisfactory in TRU for processing low-burnup targets that had been dissolved in HNO_3. A chloride-based version was investigated because the resulting transplutonium product would be in a chloride form, as is required for the Tramex process. The chloride-based Plurix process was tested successfully in glass equipment; however, in the metal equipment in TRU, the plutonium in the feed was reduced to Pu(III), which does not load on the resin. Oxidants strong enough to keep the plutonium oxidized caused excessive corrosion of equipment. Consequently, the chloride-based process was abandoned as a main-line process when the simple batch extraction method, which is discussed below, was successfully demonstrated.

During the processing of the last three SRP slugs, we were unable to recover plutonium, using the chloride-based Plurix process, because of equipment failure. A delay for repair would have caused a drastic reduction (because of radioactive decay) in the yield of einsteinium; therefore, we decided to use a simple batch extraction process after it had undergone only cursory testing. Later, we investigated flowsheet parameters in a development program.

In this batch extraction process, the feed is made 5 M in HCl, and 0.05 M HNO_3 is added as oxidant to maintain plutonium as Pu(IV). This solution is contacted with two volumes (feed solution = 1 volume) of 1 M di(2-ethylhexyl)phosphoric acid (HDEHP) in diethylbenzene (DEB) by air sparging the tank.

The aqueous phase is pumped from the tank, and the organic phase is scrubbed with three one-volume contacts of 5 M HCl--0.02 M HNO_3. The scrub solutions are added to the curium product solution. A nitrate-free wash is made with 5 M HCl, which is collected as the first fraction of the plutonium product. The nitrate is removed to prevent it from interfering with the reductant in the next step. The key to the process is the use of an organic-soluble reductant (0.25 volume of 0.2 M 2,5-di-*tert*-butylhydroquinone in 2-ethylhexanol) to reduce the extracted plutonium in the

organic phase and to allow it to be stripped into 5 M HCl (1 volume) containing 0.1 M hydroxylamine as a holding reductant.

The separation between plutonium and the transplutonium elements has been good, and recoveries have been high. Gross gamma decontamination factors for plutonium have been in the same range as those obtained by the ion exchange method (about 150).

An advantage of the batch extraction procedure over the ion exchange method is that it removes zirconium from both the plutonium and transplutonium product fractions. As much as 50 g of zirconium has been introduced into dissolver solutions by corrosion of metal equipment; this contaminant forms solids that can plug equipment lines when the product streams are evaporated to small volumes. Also, subsequent operation of the Tramex process is adversely affected if the feed contains more than about 1 g of zirconium per liter.

3.4 First-Cycle Solvent Extraction Process (Tramex)

The Tramex solvent extraction process,[7] which is used for separating the actinide and the lanthanide elements, is operational in TRU. Its performance has been satisfactory; only a few minor difficulties have been experienced. In recent runs, gross gamma decontamination factors of 175 to 200 were obtained. The curium and americium in the Tramex product are suitable for recycle to the HFIR without further decontamination because remote refabrication techniques will be used in preparing the targets.

An important process development is the modified method for adjusting the composition of the feed. Originally, when LiCl was added and the feed was heated to remove excess acid, the feed was concentrated to only 8 M LiCl, which reduced the excess acid concentration to only 0.5 M, and then it was adjusted to 11 M LiCl using 13.5 M LiCl. This gave a large feed volume. The limiting factor was the hydrolysis of zirconium when the acid concentration fell below 0.5 M. In the modified procedure the acid is completely volatilized and the zirconium is allowed to hydrolyze and precipitate while the lithium is concentrated to 12 M (137°C). Then, sufficient HCl is added to bring the acid concentration to 1 M, and the

feed is digested at 120°C for 1 hr to redissolve the hydrolyzed zirconium and to remove excess acid.

Two other changes have been made in the Tramex process recently. For unknown reasons we were not able to control the loss of acid in the feed (due to radiolysis) by the addition of methanol, a procedure which had been successful in the Curium Recovery Facility[8]. However, we found that adding concentrated HCl to the feed periodically to compensate for acid loss was a satisfactory alternative.

We have also modified the method by which we prevent the oxidation of cerium. Previously, $SnCl_2$ was added to the feed for this purpose. Currently, the organic-soluble reductant di-*tert*-butylhydroquinone (DBHQ) is included in the extractant at a concentration of 0.05 *M*. Although both methods are equally effective, the $SnCl_2$ had the disadvantage of loading the solvent, which was particularly troublesome when recycle material was processed.

In the two years that TRU has been in operation, more than 300 g of ^{244}Cm (plus associated transuranium elements) has been processed in 13 Tramex processing runs. The latest version of the Tramex flowsheet is given in Fig. 3.3. The raffinate from the plutonium recovery step is adjusted so that it is 11 *M* in LiCl and 0.2 *M* in HCl, and then it is fed to the Tramex process at the rate of 0.5 liter/hr. The transuranium elements are extracted into a solution that is 0.6 *M* in Adogen 364-HP (a high-purity tertiary amine), 0.03 *M* in HCl, and 0.05 *M* in DBHQ, in diethylbenzene (DEB) diluent. The flow rate of the extractant is 1.5 liters/hr. Small amounts of extracted lanthanides are scrubbed out of the organic phase, using a solution of 11 *M* LiCl--0.2 *M* HCl (flow rate, 0.7 liter/hr), and the actinides are stripped from the solvent, using 8 *M* HCl (flow rate, 0.5 liter/hr).

3.5 Partitioning of Actinides

A typical Tramex product obtained from the processing of irradiated ^{242}Pu targets contains up to 10,000 times as much americium and curium as the total amount of transcurium elements. The next step required in

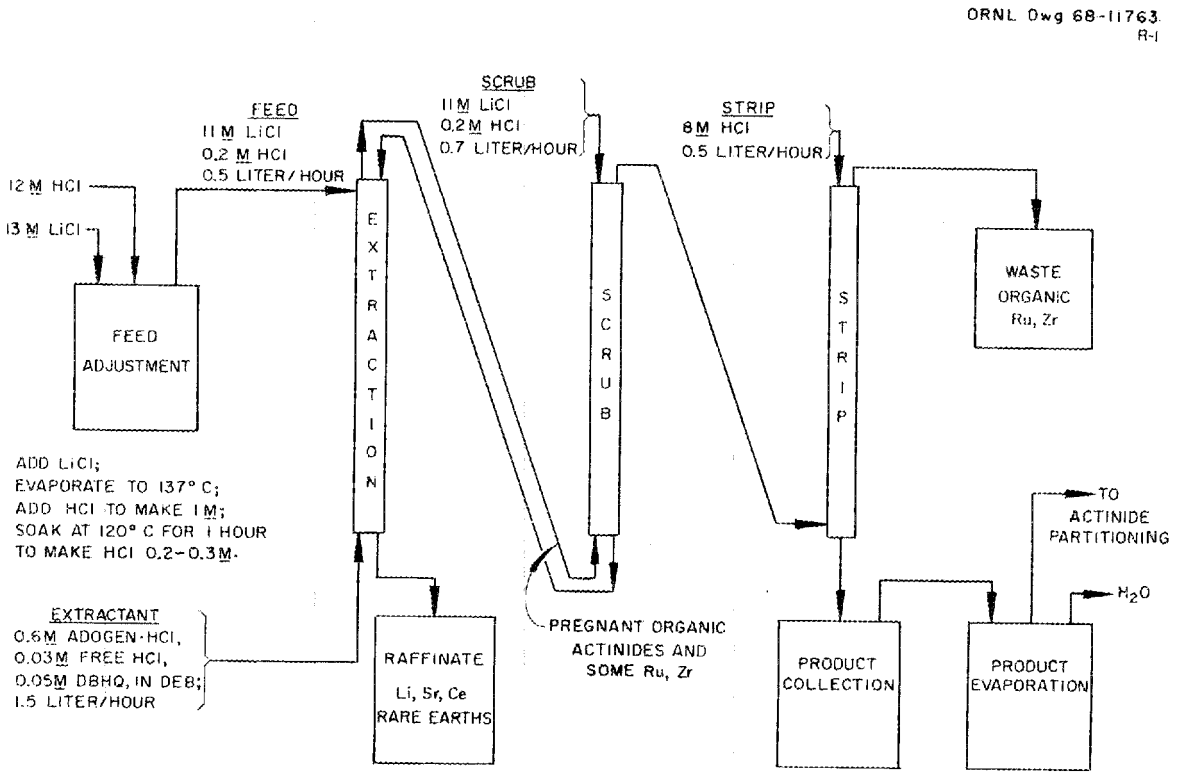

Fig. 3.3. Flowsheet for the Tramex Process for Separating Lanthanides from Actinides.

TRU, namely, the separation of the transcurium elements from americium and curium, is now being accomplished using a LiCl-based anion-exchange[9] process. This process, which had been used successfully in the laboratory for years, was scaled up in 1967 in a development facility (cell 5 in TRU) to process the Tramex product solutions from the prototype targets, the SRP slugs, and five of the SRP-HFIR targets. Then, new equipment (of glass and plastic) was installed in the TRU processing cells for use on an even larger scale. More than 200 g of ^{244}Cm, plus associated ^{243}Am and transcurium elements, has been processed using this equipment. The largest single batch contained about 35 g of ^{244}Cm and ^{243}Am. We are installing permanent equipment that has been fabricated from tantalum.

The method for preparing the feed for the LiCl-based anion exchange process has been sufficiently improved that we no longer have difficulty in handling the feed solution. In the feed preparation step, the Tramex product is contacted with 1 M HDEHP in DEB to remove zirconium and any residual organic material that might precipitate and interfere with the ion exchange process. Then the Tramex product, which is 2 to 6 M in LiCl, is concentrated until it is 13.1 M in LiCl by evaporation to a temperature of 142.5°C. Hydrochloric acid is added to bring the acid concentration to about 1 M, and the solution is digested to remove excess acid (as during the Tramex feed adjustment). This procedure circumvents the problems involving hydrolysis and precipitation, and permits the handling of up to 1 g of zirconium in a batch of feed.

The feed is passed through an anion-exchange column of Dowex 1-X10 resin (200-300 mesh). The actinides and many of the contaminants are sorbed on the anion resin from 12 M LiCl. Curium losses in the feed effluent have been less than 0.01%.

Most of the nickel and the rare earths are eluted from the resin with five to eight column displacement volumes of 10 M LiCl--0.1 M $NH_2OH \cdot HCl$--5 vol % CH_3OH. The hydroxylamine is used to maintain cerium in the trivalent state, and the methanol is used to suppress the formation of radiolytic gases. Elution with this solution is continued until an increase in alpha activity (due to ^{244}Cm) is noted in the effluent. (Curium losses to this wash have been less than 1.0%.) The eluent is then changed

to 9 M LiCl--0.1 M HCl, and elution is continued until about 90 to 95% of the curium is eluted. At this point, the eluent is changed to 8 M HCl, and the transcurium elements are stripped off the resin column. If the volume of feed is too large to be processed in a single loading of the resin column, the transcurium element fraction is recycled to the feed of the subsequent loading, and the loading-eluting cycle is repeated until all of the Tramex product solution has been processed. Then the transcurium element fraction is given a final cycle of purification, using the LiCl-based anion-exchange process. The entire quantity of transcurium elements, along with only a very small fraction of the original americium and curium, is collected in a single solution. This greatly simplifies the subsequent separation of the transcurium elements.

We are developing a continuous solvent extraction process (Hepex)[10] to separate the transcurium elements from the americium and curium. In the Pharex process, which was considered originally, the heavy actinides are separated from americium and curium by preferential extraction into 2-ethylhexyl phenylphosphonic acid (HEH[ϕP])from dilute HCl. The Pharex process can provide the required separation under ideal conditions; however, the presence of 100 ppm of zirconium in the feed increases the distribution coefficients and reduces the separation factors to intolerably low values. Thus, Pharex is not a satisfactory process for use in TRU because much of the TRU equipment is made of Zircaloy-2. The distribution coefficients in the Hepex process, in which the extractant is di(2-ethylhexyl)phosphoric acid (HDEHP), are not affected by the presence of zirconium.

3.6 Separation of Transcurium Elements

The transcurium elements are separated from each other on a high-pressure ion exchange column by chromatographic elution with α-hydroxyisobutyrate.[11] Columns of various sizes are used. The column used in recent runs was essentially a 3/8-in.-O.D., 4-ft-long stainless steel tube that was filled with about 40 ml of 20-μ beads of Dowex 50-X8 ion exchange resin in the ammonium form. The column was heated to 80°C.

The flowsheet is given in Fig. 3.4. The actinides in the transcurium elements fraction from the LiCl process were precipitated as the hydroxides and washed, to remove LiCl. Then they were redissolved in nitric acid, which was subsequently diluted to less than 0.1 M. (Nitric acid was used because the equipment is made of stainless steel.) The feed solution was pumped through the column at a rate of about 6 ml/min, which caused a pressure drop across the column of about 1000 psi. After loading, the column was washed with 50 ml of water. Then a solution of 0.3 M NH_4NO_3 was pumped through the column to replace NH_4^+ that had been displaced by excess acid in the feed. This step is essential to good chromatographic separation during the elution. Fermium and einsteinium were eluted using 0.25 M α-hydroxyisobutyrate (at a pH of 4.15) that was pumped at the rate of about 5 ml/min. The elution of the fermium-einsteinium fraction is observed as a peak of alpha activity from the einsteinium. To ensure that there was no loss of einsteinium to the californium fraction, the collection of the first product solution (i.e., the fermium-einsteinium fraction) was continued until the californium alpha peak started to appear. Then californium was collected until the alpha activity in the effluent decreased to background. The eluent was then changed to 0.5 M α-hydroxyisobutyrate (pH of 4.8) to hasten the elution of the berkelium-curium fraction.

The californium and berkelium product fractions were converted to 8 M HNO_3 solutions by cation exchange. The californium solution was a purified product. The berkelium was in a convenient form for further purification using the Berkex process. In the Berkex process, berkelium is oxidized, by the addition of 0.5 M $NaBrO_3$, to Bk(IV), which is selectively extracted into 1 M HDEHP in decane. The berkelium is then stripped from the solvent, using 8 M HNO_3—0.1 M H_2O_2, which reduces the berkelium to Bk(III).

The einsteinium and fermium were separated from the californium and purified by using additional cycles of high-pressure ion exchange treatment. Final product fractions in α-hydroxyisobutyrate solution were converted to HNO_3 solutions by using a cation exchange method.

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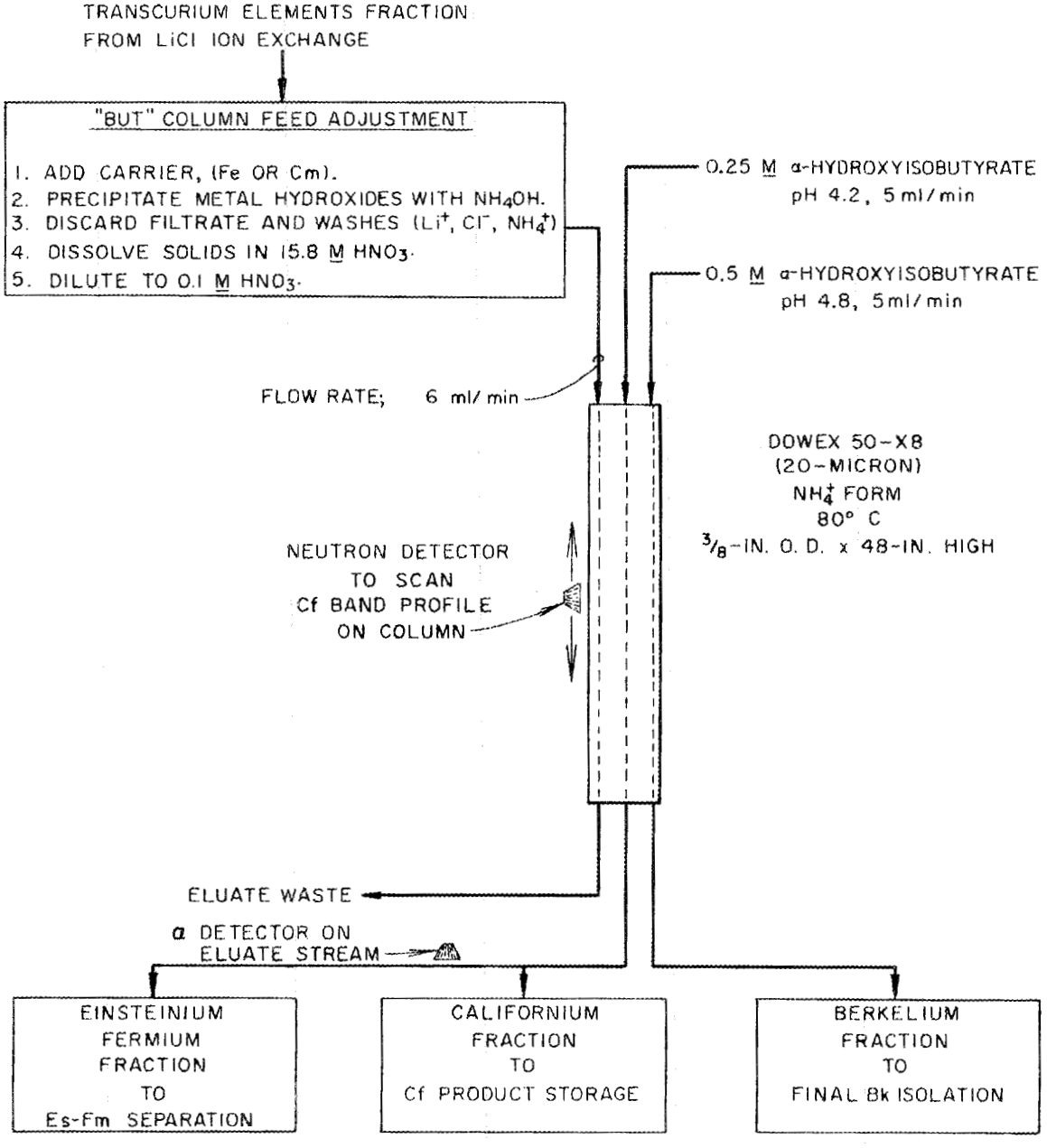

Fig. 3.4. Transuranium Element Isolation by Elution with α-Hydroxyisobutyrate from a Cation Resin Column.

3.7 Preparation of Actinide Oxides

A sol-gel process that can be operated remotely has been developed for preparing mixed ^{243}Am - ^{244}Cm oxide microspheres that are suitable for use in fabricating HFIR targets.[12] About 39 g of americium-curium oxide product in the desired size range (50-170 μ), which was prepared in Building 4507 during the development stages, has been fabricated into HFIR targets (six), and an additional 27 g is ready to be fabricated.

Equipment for the routine production of sol-gel oxide microspheres in TRU is being fabricated. In the first step of the proposed process, the nitrate solution is adjusted to 0.05 to 0.2 M HNO_3 and is then added to ammonium hydroxide that is being agitated vigorously. The resulting precipitate of hydrous oxides of americium and curium is fluidized and washed continuously with water in a conical washer. The washed precipitate is allowed to settle, and the clear supernate is removed. The remaining slurry is peptized to a stable sol by heating at 80°C for 2 hr. The sol is fed to a tapered drying column through a two-fluid nozzle. The flow rates of the sol and the second fluid (2-ethylhexanol) are adjusted to produce sol droplets of the desired size. The droplets are allowed to fall through the tapered column countercurrent to 2-ethylhexanol, which removes water from the sol and thus converts the sol droplets to gel microspheres. The gel microspheres are dried in air at 200°C and, finally, are calcined in air at 1150°C.

3.8 Fabrication of Targets

Most of the americium and curium that are recovered from irradiated targets will be incorporated into recycle targets and reirradiated in the HFIR to produce transcurium elements. The first six recycle targets have been fabricated in TRU, using the remote equipment installed in cells 1, 2, and 3. For each of the six target assemblies, actinide oxide powder, which was prepared during developmental work on the curium sol-gel process, was blended with aluminum powder, and the resulting mixture was pressed

into aluminum-clad pellets. The pellets were inspected to ensure conformity to size and weight requirements and heated under vacuum at 400°C to remove the die lubricant. From 34 to 36 pellets (total length, 20 ± 0.5 in.) were loaded into a target tube that had previously had a bottom end cap welded in place. A top support liner, cut to the proper length for the target, was inserted. The tube was evacuated and filled with helium; then the top end cap was installed and welded to the tube. The tube was examined for leaks in a helium leak detector; was cleaned, dried, and smeared for contamination; and was x-rayed to ensure the integrity of the welds and to determine that the end spacers and pellets were in place. The tube was pressed hydrostatically (at 10 tsi) to make a good heat transfer bond between the pellets and the tube, and the tube was helium leak-tested again. Finally, the hexagonal coolant-flow tube was mechanically staked to fins on the target rod, and the finished target assembly was cleaned and transferred to the HFIR.

Although numerous minor problems have been encountered, this equipment is considered to be fully operational.

3.9 Corrosion of Zircaloy-2

We found that the corrosion rate of Zircaloy-2 equipment was unexpectedly catastrophic when certain mixtures of HCl and HNO_3 were used, and that the rate at which HCl corrodes Zircaloy-2 in a radiation environment was much higher than we had anticipated. We made some corrosion tests to study these situations.

Zircaloy-2 was not attacked significantly by acid mixtures that were less than 2 M in HCl. In 3 M HCl the corrosion rate appeared to be significant ($\sim$ 5 mils/year) when the concentration of HNO_3 was as low as 0.5 M, and was definitely catastrophic (75 to 200 mils/year) at 1 to 6 M HNO_3. Further, the attack was not uniform. In all HCl-HNO_3 mixtures in which the HCl concentration was 4 M or greater and the HNO_3 concentration was greater than a few tenths molar, the corrosion was catastrophic and the attack was not uniform.

The Zircaloy-2 corrosion rate in 6 M HCl at a solution power density of 37 w/liter (13.5 g ^{244}Cm/liter) was constant at about 60 mils/year. The data for a solution power density of 8 w/liter indicate that the corrosion rate was increasing throughout the test period. After 24 hr the rate was 1.3 mils/year; during the next 42.3 hr it was 14 mils/year; during the next 94 hr it was 18 mils/year; and during the final 46 hr it was 32 mils/year. This compares with corrosion rates of only a few mils per year in nonradioactive HCl solutions.

3.10 HFIR Target Rod Failures

Sixteen TRU target rods developed longitudinal cracks in the cladding after irradiation in the HFIR for 125 to 175 days at a power level of 100 Mw. These rods were from a group of 17 that had been irradiated prior to this time, for about one year in a Savannah River Plant reactor.

The Metals and Ceramics Division began an extensive program to determine the mechanisms that caused the failures and to develop an improved target design. Details of their efforts to date have recently been reported.[13] The nature of the problem precludes rapid solution; proof of a new target design will require a minimum of 18 months of irradiation in the HFIR. Because of our vital interest in this matter, the status of the work is summarized here.

Various techniques were used in analyzing the failure: visual inspection, measurement of dimensional changes, metallography, vacuum-fusion gas analysis, determination of mechanical properties, electron microscopy, and x-ray diffraction.

It was concluded that exposure of the aluminum alloy cladding material to the high fast-neutron flux in the HFIR causes a rapid decrease in the ductility. Fission products accumulate and fill the void volume in the pellets, and the gases increase the pressure in the remaining voids. This results in a gradual increase in the stresses in the cladding. When the stresses reach the yield point of the cladding, the embrittled cladding fractures instead of yielding.

We have formulated an engineering model to guide the development of an improved design. Figure 3.5 summarizes the results of calculations

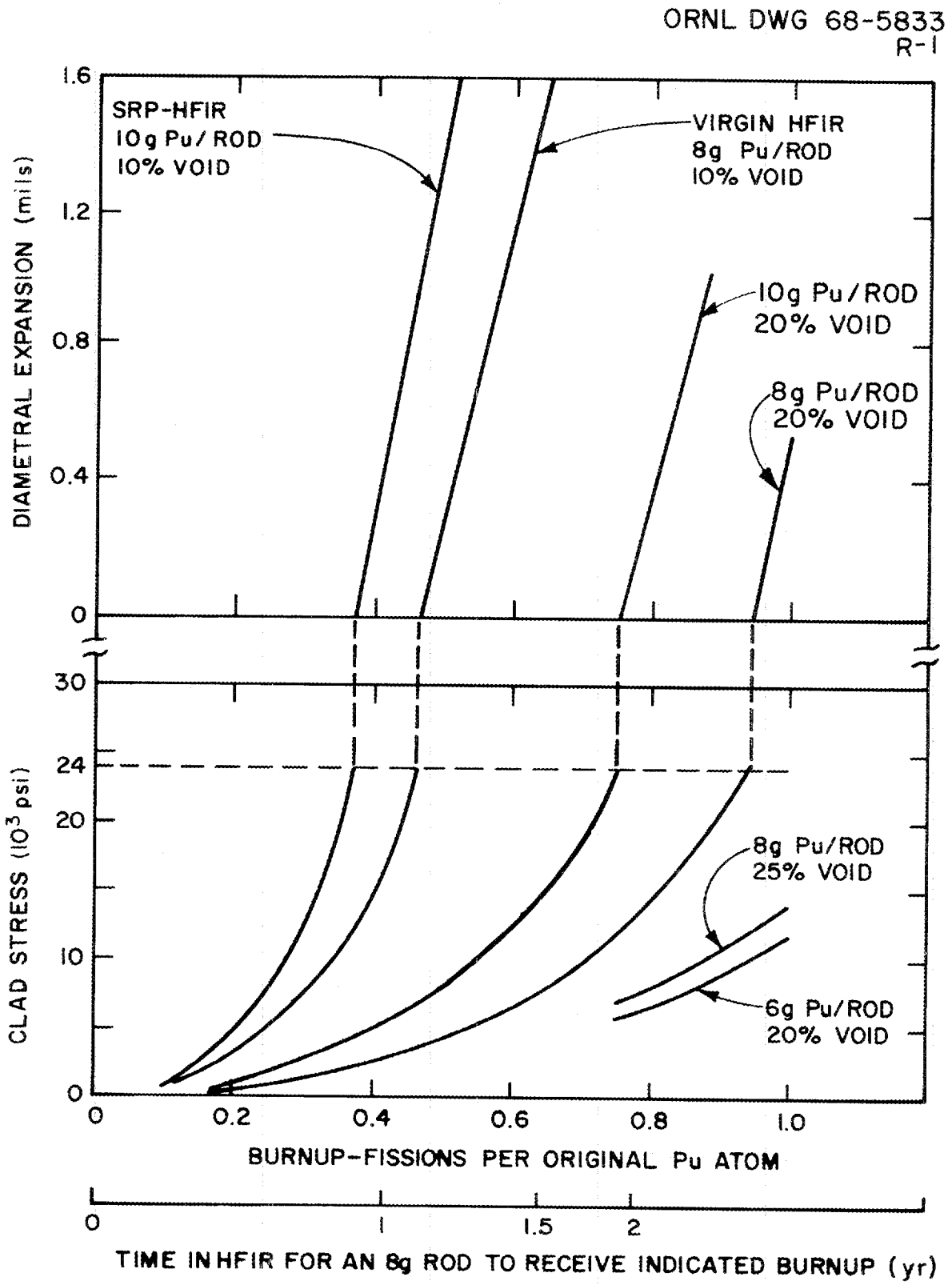

Fig. 3.5. Effect of Burnup and Fabrication Variables on Clad Stress.

that have been made. The lower family of curves shows the stress in the cladding of various targets as a function of the burnup. The abscissa is the number of fissions occurring per atom of plutonium in the original target rod. The dashed line at 24,000 psi indicates the probable stress at which the SRP-HFIR targets failed. This value, which was determined from specimens of the aluminum coolant-flow tubes that were on the targets, was used as a typical stress at which the cladding on any target might fail. The upper graph has as its ordinate the diametral expansion that would occur if the target sheath yielded (instead of rupturing).

The curve for the SRP-HFIR targets (10 g of ^{242}Pu per target and 10% void space in the pellets) is in fair agreement with the failure data. The model indicates failure at 0.375 fission per original plutonium atom; the targets actually failed at burnups of from 0.23 to 0.37 fission per original plutonium atom. (The early failures might be attributed to non-uniform loading and variation in pellet densities.) The curve for "virgin HFIR targets containing 8 g of ^{242}Pu per target and having 10% void volume" represents a group of 13 targets that have recently been removed from the HFIR for processing. It indicated that these targets would fail at a burnup of 0.46 fission per original plutonium atom. However, inspection of the targets, which had been irradiated to burnups of from 0.48 to 0.52 fission per original plutonium atom, disclosed no failures.

Each of the three curves labeled "8 g plutonium/rod, 20% void," "8 g plutonium/rod, 25% void," and "6 g plutonium/rod, 20% void" represents pairs of special targets that were fabricated during the past year (before the mathematical model was formulated) and that are now being irradiated to determine whether smaller loading and/or pressing of the pellets to lower densities (and larger void volumes) will result in longer target lifetimes. Calculations indicate that none of these targets will fail prior to the time that the irradiations are scheduled to be terminated. If this proves to be true, the failure problem will be resolved, for all practical purposes, in a way that does not restrict future production. Of course, there will still be great interest in the aluminum embrittlement phenomenon; therefore, further studies will be made to elucidate it.

The curve labeled "10 g plutonium/rod, 20% void" represents a group of targets that will be fabricated in the near future from our remaining stock of plutonium oxide. The expected failure point, 0.75 fission per original plutonium atom, is well beyond the proposed irradiation time.

4. SPECIAL PROJECTS

Numerous projects that are being carried out by various groups at ORNL and at other sites require the services available at TRU for the preparation of special materials, which are not normally produced in mainline efforts. The phases of these projects involving activities at TRU are reported here; the end results of the research are reported elsewhere.

4.1 Californium Targets

Three special californium targets were fabricated and irradiated for the primary purpose of producing einsteinium. A second purpose of the irradiations was to refine some of the calculated values of the cross sections in the chain of isotopes above californium. Surprisingly, we learned that the capture cross section of ^{252}Cf is considerably higher than previously supposed, apparently in the range of 40-50 barns instead of 7-10 barns. Additional irradiation experiments will be necessary to further refine these measurements. Because of the high cross section, the irradiation times used were far too long for optimum einsteinium production and thus a significant fraction of the californium was consumed in nonproductive reactions. About 6 μg of einsteinium, as compared with the 18 μg expected, was recovered for shipment to customers. Other special materials, including 1 mg of ^{244}Pu, 26 μg of ^{248}Cm, and 12 aluminum alloy tensile specimens, were irradiated with the californium in these targets. The tensile specimens were used for studying the target failure problem.

4.2 Fabrication of a Neutron Source Using Californium

The first significant neutron source using californium was fabricated in January 1968 for fast-neutron activation studies in the Transuranium Research Laboratory. About 350 μg of ^{252}Cf (8×10^8 neutrons/sec) was loaded into a small aluminum source capsule, 3/8 in. in diameter by about 1 in. long, using a new technique. The californium was precipitated as the hydroxide, sorbed on aluminum powder, and retained on a pressed aluminum powder filter disk at the bottom of a standard target pellet liner. The liner and the powder matrix were dried and pressed in the usual manner, and then the completed pellet was welded into an outer aluminum capsule.

4.3 Uranium Irradiations

Assistance was provided in the preparation and irradiation of 50-mg quantities of U_3O_8 (42 mg of ^{236}U) in HFIR rabbits for the production of ^{237}U. This was the first productive use of the HFIR hydraulic rabbit facility. The rabbits were subsequently processed by personnel at the Los Alamos Scientific Laboratory, and the uranium was used in an underground nuclear event for the measurement of the fission cross section of ^{237}U.

4.4 Einsteinium Irradiations

Rabbits containing microgram amounts of ^{253}Es were fabricated and were irradiated for three days in the HFIR to produce 39.3-hr ^{254m}Es. The irradiated rabbits were disassembled in TRU after a very short cooling period, and the pellets containing the einsteinium were shipped by air to Argonne National Laboratory. The aluminum rabbit tube had to be removed prior to shipment because it was a strong source of penetrating radiation (from neutron activation products of aluminum).

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6. APPENDIX

We have tabulated the decay data and the cross section data that we use in planning irradiation-processing cycles, calculating production forecasts, and assaying products. The tables will be reproduced completely in each of these semiannual reports, and changes made since the preceding report will be indicated. We wish to state clearly that these data merely represent numbers being used in our calculations and that the data are presented on a "best efforts" basis. Although the information is intended to be definitive, it has not been checked and cross-checked sufficiently to be considered "publishable."

The Transplutonium Element Production Program is now making nuclides available in increasing abundance and purity; therefore, in the next few years we anticipate a burgeoning literature concerning nuclear constants for the transuranium nuclides. However, since we need such data at the present time, it will not be feasible for us to wait until highly reliable sources, such as Lederer[14] and Wapstra[15], can publish data that have been fully evaluated.

We welcome telephone calls to point out errors or indicate additional sources of information. Please contact John Bigelow, FTS 615-483-1872 or, by commercial telephone service, 615-483-8611, ext. 3-1872.

Decay Data

Table A-1 is a list of all nuclides of interest to the Transplutonium Element Production Program, (i.e., all that can be produced by neutron bombardment of ^{238}U). The list includes values for half-lives and branching ratios or partial decay half-lives, along with literature references, where available. The system of references is that used by the Nuclear Data Project here at ORNL in their widely distributed "Nuclear Data Sheets."

The references used in Table A-1 are decoded in Table A-2. Table A-3 lists derived data, such as specific activities, along with information concerning the hazard associated with handling these nuclides.

Table A-1. Half-Life Values[a] for Isotopes of Transuranium Elements

Nuclide	Total Half-Life	Reference[b]	Partial Half-Life for Spontaneous Fission	Reference[b]
^{237}Np	$(2.14 \pm 0.01) \times 10^{6}$ y	60Br12	$>10^{18}$ y	61Dr4
^{238}Np	2.10 d	50F53		
^{239}Np	2.339 ± 0.010 d	59C93		
^{240}Np	63 ± 2 m	60Le3		
^{240m}Np	7.3 ± 0.3 m	48H61		
^{241}Np	16 m	60Le3		
^{241m}Np	3.4 h	60Le3		
^{238}Pu	87.60 ± 0.06 y	59M126	$(5 \pm 0.6) \times 10^{10}$ y	61Dr4
^{239}Pu	24,413 ± 30 y	51I3	5.5×10^{15} y	52S67
^{240}Pu	6,580 ± 40 y	51I3	$(1.340 \pm 0.015) \times 10^{11}$ y	62Wa13
^{241}Pu	14.2 ± 0.2 y			
^{242}Pu	$(3.869 \pm 0.016) \times 10^{5}$ y		$(7.45 \pm 0.17) \times 10^{10}$ y	63Ma50
^{243}Pu	4.98 ± 0.02 h	53E8		
^{244}Pu	$(8.28 \pm 0.10) \times 10^{7}$ y		$(6.55 \pm 0.32) \times 10^{10}$ y	
^{245}Pu	10.6 ± 0.4 h	56B92		
^{246}Pu	10.85 ± 0.02 d	56N23		
^{241}Am	432.5 ± 1.0 y		$(2.3 \pm 0.8) \times 10^{14}$ y	61Dr4
^{242}Am	16.01 ± 0.2 h	53K38		
^{242m}Am	152 ± 7 y	59B221		
^{243}Am	7340 ± 50 y			
^{244}Am	10.1 ± 0.1 h	62Va8		
^{244m}Am	26 m	50S61		
^{245}Am	2.07 ± 0.02 h	56B92		
^{246}Am	25.0 ± 0.2 m	55E16		
^{246m}Am	40 ± 7 m	67Or02		
^{247}Am	24 ± 3 m	67Or02		
^{242}Cm	162.7 ± 0.1 d	57P52	7.2×10^{6} y	51H87
^{243}Cm	32 y	57A70		
^{244}Cm	18.11 ± 0.07 y	63Be54	$(1.346 \pm 0.006) \times 10^{7}$ y	65Me02
^{245}Cm	9600 ± 290 y	61Ca1		
^{246}Cm	5640 ± 140 y	54F19	$(1.66 \pm 0.10) \times 10^{7}$ y	65Me02
^{247}Cm	$(1.64 \pm 0.20) \times 10^{7}$ y	63Fi08		
^{248}Cm	$(4.2 \pm 0.3) \times 10^{5}$ y	56B91	$(4.6 \pm 0.5) \times 10^{6}$ y	56B91
^{249}Cm	64 ± 3 m	58E06		
^{250}Cm	$(1.74 \pm 0.24) \times 10^{4}$ y	66RG01	$(1.74 \pm 0.24) \times 10^{4}$ y	66RG01
^{249}Bk	314 d	57E01	$>1.4 \times 10^{9}$ y	57E01
^{250}Bk	3.222 ± 0.005 h	59V02		
^{251}Bk	57 ± 1.7 m	66RG04		

Table A-1 (continued)

Nuclide	Total Half-Life	Reference[b]	Partial Half-Life for Spontaneous Fission	Reference[b]
^{249}Cf	323 y		~4.5 x 10^8 y	57E01
^{250}Cf	13.2 ± 0.5 y	65Me02	(1.73 ± 0.06) x 10^4 y	62Ph2
^{251}Cf	892 y			
^{252}Cf	2.646 ± 0.004 y	65Me02	85.5 ± 0.5 y	65Me02
^{253}Cf	17.6 ± 0.3 d	65Me02		
^{254}Cf	60.5 ± 0.2 d	63Ph01	60.5 ± 0.2 d	63Ph01
^{253}Es	20.7 ± 0.3 d	66RG01	(6.3 ± 0.2) x 10^5 y	65Me02
^{254}Es	276 d	67Un01	~2.5 x 10^7 y	
^{254m}Es	39.3 ± 0.2 h	62Un1		
^{255}Es	39.8 ± 1.2 d	66RG01	2440 ± 140 y	
^{256}Es	<1-2 h	55C30		
^{254}Fm	3.24 ± 0.01 h	56J09	228 ± 1 d	
^{255}Fm	20.07 ± 0.07 h	64As01	(1.0 ± 0.6) x 10^4 y	
^{256}Fm	2.7 ± 0.1 h	65Si14	2.7 ± 0.1 h	65Si14
^{257}Fm	94 ± 10 d	66RG01		
^{258}Fm	~2s		~2s	

[a]The half-life values used in this table were being used at TRU on July 1, 1968.

[b]References are decoded in Table A-2.

Table A-2. References for Table A-1

Code	Reference
48H61	E. K. Hyde, M. H. Studier, and W. M. Manning, ANL-4143 (April 15, 1948) and ANL-4182 (August 4, 1948).
50F53	M. S. Freedman, A. H. Jaffey, and F. Wagner, Jr., Phys. Rev., 79, 410-411 (1950).
50S61	K. Street, Jr., A. Ghiorso, and G. T. Seaborg, Phys. Rev., 79, 530-531 (1950).
51H87	G. C. Hanna, B. G. Harvey, N. Moss, and P. R. Tunnicliffe, Phys. Rev., 81, 466-467 (1951).
51I73	M. G. Inghram, D. C. Hess, P. R. Fields, and G. L. Pyle, Phys. Rev., 83, 1250 (1951).
51S55	J. C. Sullivan, G. L. Pyle, M. H. Studier, P. R. Fields, and W. M. Manning, Phys. Rev., 83, 1267-1268 (1951).
52S87	E. Segrè, Phys. Rev., 86, 21-28 (1952).
53E8	D. W. Engelkemeir, P. R. Fields, and J. R. Huizenga, Phys. Rev., 90, 6 (1953).
53K38	T. K. Keenan, R. A. Penneman, and B. B. McInteer, J. Chem. Phys., 21, 1802-1803 (1953).
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55E16	D. Engelkemeir, P. R. Fields, T. Fried, G. L. Pyle, C. M. Stevens, L. B. Asprey, C. I. Browne, H. Louise Smith, and R. W. Spence, J. Inorg. Nucl. Chem., 1, 345-351 (1955).
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56H23	D. C. Hoffman and C. I. Browne, J. Inorg. Nucl. Chem., 2, 209 (1956).
56J09	M. Jones, R. P. Schuman, J. P. Butler, G. Cowper, T. A. Eastwood, and H. G. Jackson, Phys. Rev., 102, 203-207 (1956).
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58E06	T. A. Eastwood and R. P. Schuman, J. Inorg. Nucl. Chem., 6, 261-262 (1958).
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60Br12	F. P. Brauer, R. W. Stromatt, J. D. Ludwick, F. P. Roberts and W. L. Lyon, J. Inorg. Nucl. Chem., 12, 234-235 (1960).
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62Va8	S. E. Vandenbosch and P. Day, Nucl. Phys., 30, 177-190 (1962).
62Wa13	D. E. Watt, F. J. Bannister, J. B. Laidlaw, and F. Brown, Phys. Rev., 126, 264-265 (1962).
62Be34	W. C. Bentley, as reported by D. Metta, H. Diamond, R. F. Barnes, J. Milsted, J. Gray, Jr., D. J. Henderson, and C. M. Stevens, J. Inorg. Nucl. Chem., 27, 33-41 (1965).
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64As01	F. Asaro, S. Bjrnholm, and I. Perlman, Phys. Rev., 133, B291-B300 (1964).
65Me02	D. Metta, H. Diamond, R. F. Barnes, J. Milsted, J. Gray, Jr., D. J. Henderson, and C. M. Stevens, J. Inorg. Nucl. Chem., 27, 33 (1965).
65Si14	T. Sikkeland, A. Ghiorso, R. Latimer, and A. E. Larsh, Phys. Rev., 140, B227 (1965).
66kC01	Combined Radiochemistry Group, LRL, LASL, and ANL, Phys. Rev., 148 No. 3, 1192-1198 (1966).
66RG04	Argonne Heavy Element Group (unpublished data).
67Or02	C. J. Orth, W. R. Daniels, B. H. Erkkila, E. O. Lawrence, and D. C. Hoffman, Phys. Rev. Letters, 19, No. 3, 128-131 (1967).
67Un01	J. Unik, private communication to P. Fields (1967).

Table A-3. Properties[a] of Transuranium Nuclides

Nuclide	Half-life	Energies of Prin. Emissions (Mev) α	β	Specific Activity (Ci)	(w/g)	(α cpm/mg[c])	(β dpm/mg)	(Neutrons min^{-1} mg^{-1})	Hazard[b] $MPC_{a(40)}$ (μCi/cm^3)	Body Burden (μCi)	(μg)
^{237}Np	2.14×10^6 y	4.78		7.07×10^{-4}	2.07×10^{-5}	8.01×10^5			4×10^{-12}	0.06	84.9
^{238}Np	2.10 d		0.25 1.24	2.61×10^5	1.27×10^3		5.80×10^{14}				
^{239}Np	2.359 d		0.332 0.427	2.32×10^5	5.86×10^2		5.14×10^{14}		8×10^{-7}	30	1.29×10^{-4}
^{240}Np	63 m		0.89	1.24×10^7	1.03×10^5		2.76×10^{16}				
^{240m}Np	7.3 m		2.18 1.6	1.07×10^8	5.33×10^5		2.38×10^{17}				
^{241}Np	16 m			4.86×10^7			1.08×10^{17}				
^{241m}Np	3.4 h			3.82×10^6			8.49×10^{15}				
^{238}Pu	87.60 y	5.49		17.2	0.569	1.94×10^{10}		148	2×10^{-12}	0.04	2.32×10^{-3}
^{239}Pu	24,413 y	5.15		6.13×10^{-2}	1.923×10^{-3}	6.94×10^7			2×10^{-12}	0.04	0.654
^{240}Pu	6,580 y	5.16		0.227	7.95×10^{-3}	2.57×10^8		66	2×10^{-12}	0.04	0.176
^{241}Pu	14.2 y	4.9	0.02	1.05×10^2	3.72×10^{-3}	3.4×10^6	2.32×10^{11}		9×10^{-11}	0.9	8.57×10^{-3}
^{242}Pu	3.869×10^5 y	4.90		3.82×10^{-3}	1.13×10^{-4}	4.32×10^6		124	2×10^{-12}	0.05	13.0
^{243}Pu	4.98 h		0.49 0.58	2.59×10^6	3.33×10^3		5.75×10^{15}		2×10^{-6}	7.2	2.78×10^{-6}
^{244}Pu	8.28×10^7 y	4.587		1.77×10^{-5}	4.93×10^{-7}	2.00×10^4		374	2×10^{-12}	0.04	2.23×10^3
^{245}Pu	10.6 h			1.21×10^6			2.68×10^{15}		2×10^{-7}	3.0	4.03×10^5
^{246}Pu	10.85 d		0.15	4.91×10^4	66.9		1.09×10^{14}				
^{241}Am	432.5 y	5.48		3.42	0.115	3.89×10^9		0.04	6×10^{-12}	0.1	0.029
^{242}Am	16.01 h		0.63 0.67	8.11×10^5	2.08×10^3		1.80×10^{15} (ref. d)		4×10^{-8}	0.06	7.39×10^{-8}
^{242m}Am	152 y	5.207	I. I.	9.73	2.92×10^{-2}	5.24×10^7	2.16×10^{10} (ref. e)		6×10^{-12}	0.07	7.19×10^{-3}
^{243}Am	7340 y	5.27		0.200	6.43×10^{-3}	2.27×10^8			6×10^{-12}	0.05	0.25
^{244}Am	10.1 h		0.387	1.27×10^6	8.74×10^3		2.82×10^{15}		2×10^{-7}	0.18	1.42×10^{-7}
^{244m}Am	26 m		1.5	2.96×10^7	8.98×10^4		6.58×10^{16} (ref. f)		4×10^{-6}	0.18	6.08×10^{-9}
^{245}Am	2.07 h		0.91	6.17×10^6	1.20×10^4		1.37×10^{16}		3×10^{-6}	12	1.94×10^{-6}
^{246}Am	25 m		1.31	3.06×10^7	2.48×10^5		6.79×10^{16}				
^{246m}Am	40 m			1.91×10^7			4.24×10^{16}				
^{247}Am	24 m			3.17×10^7			7.04×10^{16}				
^{242}Cm	162.7 d	6.11		3.32×10^3	122	3.76×10^{12}		1.20×10^6	1×10^{-10}	0.05	6.27×10^{-7}
^{243}Cm	32 y	5.79		45.9	1.677	5.20×10^{10}	3.27×10^8		6×10^{-12}	0.09	1.96×10^{-3}
^{244}Cm	18.11 y	5.80		81.1	2.832	9.18×10^{10}		6.68×10^5	9×10^{-12}	0.1	1.23×10^{-3}
^{245}Cm	9600 y	5.36		0.152	5.07×10^{-3}	1.72×10^8			5×10^{-12}	0.04	0.263
^{246}Cm	5640 y	5.4		0.258	8.45×10^{-3}	2.92×10^8		5.03×10^5	5×10^{-12}	0.05	0.194
^{247}Cm	1.64×10^7 y			8.83×10^{-5}	2.8×10^{-6}	1.00×10^5			5×10^{-12}	0.04	453
^{248}Cm	4.2×10^5 y	5.05		3.43×10^{-3}	4.78×10^{-3}	3.53×10^6		2.32×10^6	6×10^{-13}	0.01	2.91
^{249}Cm	64 m		0.9	1.18×10^7	2.064×10^4		2.62×10^{16}		8×10^{-6}	0.8	6.78×10^{-8}
^{250}Cm	1.74×10^4 y			8.20×10^{-2}	0.1			6.50×10^8			
^{249}Bk	314 d	5.4	0.125	1.67×10^3	0.358	4.16×10^7	3.71×10^{12}	7.92×10^3	9×10^{-10}	0.7	419
^{250}Bk	3.22 h		0.23	3.89×10^6	2.75×10^4		8.63×10^{15}		1×10^{-7}	0.04	1.03×10^{-7}
^{251}Bk	57 m			1.32×10^7			2.92×10^{16}				

Table A-3 (continued)

Nuclide	Half-Life	Energies of Prin. Emissions (Mev) α	β	Specific Activity (Ci)	(w/g)	(α dpm/mg[c])	(β dpm/mg)	(Neutrons min^{-1} mg^{-1})	Hazard[b] $MPC_{a(40)}$ ($\mu Ci/cm^3$)	Body Burden (μCi)	(μg)
^{249}Cf	323 y	5.81		4.45	0.166	5.03×10^9		$\sim 2 \times 10^3$	2×10^{-12}	0.04	8.98×10^{-3}
^{250}Cf	13.2 y	6.03		108	4.02	1.22×10^{11}		2.12×10^8	5×10^{-12}	0.04	3.70×10^{-4}
^{251}Cf	800 y			1.78	6.51×10^{-2}	2.01×10^9			2×10^{-12}	0.04	2.25×10^{-2}
^{252}Cf	2.646 y	6.11		536	39.0	5.88×10^{11}		1.40×10^{11}	2×10^{-11}	0.04	7.46×10^{-5}
^{253}Cf	17.6 d		0.27	2.94×10^4	14.1		6.52×10^{13}		8×10^{-10}	0.04	1.36×10^{-6}
^{254}Cf	60.5 d	5.84		8.51×10^3	1.06×10^4			7.37×10^{13}	5×10^{-12}	0.01	1.18×10^{-6}
^{253}Es	20.7 d	6.63		2.50×10^4	996	2.83×10^{13}		1.95×10^7	7×10^{-10}	0.04	1.60×10^{-7}
^{254}Es	276 d	6.42		1.86×10^3	71.9	2.11×10^{12}		8.39×10^7	2×10^{-11}	0.02	1.08×10^{-5}
^{254m}Es	39.3 h		0.48	3.14×10^5	1.18×10^3		6.97×10^{14}		5×10^{-9}	0.02	6.37×10^{-8}
^{255}Es	39.8 d			1.29×10^4			2.86×10^{13}		6×10^{-10}	0.04	3.10×10^{-6}
^{256}Es	<1-2 h			$>8 \times 10^6$			$>2 \times 10^{16}$				
^{254}Fm	3.24 h	7.20		3.81×10^6	1.68×10^5	4.31×10^{15}		2.09×10^{11}	6×10^{-8}	0.02	5.25×10^{-9}
^{255}Fm	20.07 h	7.03		6.13×10^5	2.79×10^4	6.94×10^{14}		1.29×10^9	2×10^{-8}	0.04	6.53×10^{-8}
^{256}Fm	2.7 h			4.55×10^6	5.85×10^6			4.31×10^{16}	2×10^{-9}	0.01	2.20×10^{-9}
^{257}Fm	94 d			5.41×10^3	~ 200	6.12×10^{12}					
^{258}Fm	~ 2 s			$\sim 2 \times 10^{10}$	$\sim 3 \times 10^{10}$			$\sim 2 \times 10^{20}$			

[a] The values for properties included in this table are those in use at TRU on July 1, 1968.

[b] From ICRP Publication 2, "Report of Committee II on Permissible Dose for Internal Radiation (1959)" and the 1962 Supplement.

[c] Counting geometry, 51%.

Neutron Cross-Section Data

The values of neutron cross sections used to compute transmutations in HFIR target irradiations are listed in Table A-4. This table shows six parameters describing the neutron interactions. The first is the thermal-neutron capture cross section, and the third is the neutron capture resonance integral. The second parameter is a constant that is a function of the target geometry; it is used to estimate the resonance self-shielding effect. The effective capture cross-section, σ^c_{eff}, would be:

$$\sigma^c_{eff} = \sigma^c_{2000} + \frac{\phi_{res}}{\phi_{2200}} \frac{RI}{\sqrt{1 + CN}} ,$$

where σ^c_{2200} is the thermal-neutron capture cross section, N is the number of grams of the particular nuclide in one target rod, ϕ_{res} is the average flux per unit lethargy width in the resonance region, and ϕ_{2200} is the equivalent flux of 2200-m/sec neutrons that would give the same reaction rate with a 1/v absorber as would the actual reactor flux. The effective cross section for fission is computed by a similar relationship among the last three parameters.

These cross sections are to be regarded as a self-consistent set, whereby one can compute overall transmutation effects, and as a set of arbitrary constants to be used to obtain the best fit to our data. Hopefully, these numbers and the cross sections experimentally measured on pure isotopes will agree; however, we will not allow the possibility of a discrepancy to confine us.

It should be pointed out that ^{244c}Am is a fictitious isotope that combines the properties of ^{244m}Am and ^{244g}Am, according to their relative rates of production from ^{243}Am. The use of this isotope simplifies the calculations of the main transmutation chain involving ^{244}Am.

Table A-4. Neutron Cross Sections Used to Compute Transmutations in HFIR Target Irradiations (July 1, 1968)

		Capture			Fission		
Nuclide	Half-Life	2200-m/s Cross Section (barns)	Resonance Self-Shielding Constant	Resonance Integral (barns)	2200-m/s Cross Section (barns)	Resonance Self-Shielding Constant	Resonance Integral (barns)
^{238}Pu	87.6 y	500	0	150	17.5	0	25
^{239}Pu	24,413 y	265.7	0	195	742.7	0	324
^{240}Pu	6,580 y	281	0	8453	0	0	0
^{241}Pu	14.2 y	382.9	0	166	1006.1	0	541
^{242}Pu	382,500 y	18.5	7.409	1280	0	0	0
^{243}Pu	4.98 h	80	0	0	210	0	0
^{244}Pu	8.28×10^7 y	1.6	0	0	0	0	0
^{245}Pu	10.6 h	277	0	0	0	0	0
^{246}Pu	10.85 d	0	0	0	0	0	0
^{243}Am	7,650 y	75	2.126	1500	0	0	0
^{244c}Am[a]	27.3 m	0	0	0	130	0	0
^{245}Am	2.07 h	0	0	0	0	0	0
^{246}Am	25 m	0	0	0	0	0	0
^{244}Cm	18.11 y	10	6.9	650	2.7	6.9	72
^{245}Cm	9,600 y	235	0	500	2170	0	0
^{246}Cm	5,640 y	5	0.866	85	0	0	0
^{247}Cm	1.64×10^7 y	190	0	0	260	0	0
^{248}Cm	420,000 y	5.2	1.447	250	0	0	0
^{249}Cm	64 m	50	0	0	50	0	0
^{250}Cm	17,400 y	2	0	0	0	0	0
^{249}Bk	314 d	900	0	0	0	0	0
^{250}Bk	193.3 m	350	0	0	1550	0	0
^{249}Cf	323 y	270	0	0	1600	0	0
^{250}Cf	13.2 y	1000	0	0	0	0	0
^{251}Cf	800 y	1600	0	0	3000	0	0
^{252}Cf	2.646 y	4	0	0	0	0	0
^{253}Cf	17.6 d	5.2	0	0	0	0	0
^{254}Cf	60.5 d	2	0	0	0	0	0
^{253}Es	20.7 d	345	0	0	0	0	0
^{254m}Es	39.3 h	1.26	0	0	1840	0	0
^{254}Es	276 d	20	0	0	3060	0	0
^{255}Es	39.8 d	40	0	0	0	0	0
^{254}Fm	3.24 h	100	0	0	0	0	0
^{255}Fm	20.07 h	100	0	0	100	0	0
^{256}Fm	2.7 h	20	0	0	0	0	0
^{257}Fm	94 d	100	0	0	100	0	0

[a] To simplify calculations we use a fictitious isotope, ^{244c}Am, which combines the properties of ^{244m}Am and ^{244g}Am according to their relative rates of production from ^{243}Am.

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